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June 29, 1988

Dr. R.E. Miller
Office of Naval Research
Code 1132P
800 N. Quincy Street
Arlington, VA 22217-5000

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Dear Dr. Miller:

We are writing to provide you with a brief annual report for the end of the first year of our contract with you, N00014-87-K-0546, "Selective Dissociation Pathways Induced by Local Electronic Excitation of Molecules in the Gas Phase and Adsorbed on Surfaces." We are happy to report that the construction of the new high resolution crossed laser molecular beam apparatus required for the proposed studies is going as well as we had planned in our "End of Fiscal Year" report to the Mechanics Division in the beginning of October. Details follow.

Our scientific goal for the three years of our grant is to investigate how local electronic excitation of a polyatomic molecule can result in selective decomposition channels over other energetically allowed ones. In the proposed work, we study the generality of using direct excitation to an excited potential energy surface repulsive in a specific bond to induce fission of that bond, an excitation found to be selective in CH_2BrI photodecomposition. In addition we investigate two new excitation mechanisms for controlling chemical fragmentation, the role of triplet states in promoting fission to radical products over molecular elimination and the efficacy of using electronic excitation localized on a particular functional group to favor dissociation at or near that group. The experimental technique utilized, measurement of photofragment velocity and angular distributions by the cross laser-molecular beam time-of-flight technique, incorporates specific capabilities necessary for identifying all the primary dissociation channels resulting from local electronic excitation. The work is fundamental to understanding and controlling reactions of isolated polyatomic molecules both in the gas phase and adsorbed on surfaces.

As stated in our proposal on Page 28, the first year and a half of the grant period involves the dedicated construction of the high resolution crossed laser molecular beam time-of-flight apparatus for these studies. The progress of this construction and outline for the next

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year follows. The final machining of the source and main chamber of the apparatus was completed by the Central Shop of the University of Chicago in early February 1988. Since then we have installed the source and main chambers in the laboratory, integrated the mechanical and diffusion pumps and differential region turbomolecular pump, incorporated the necessary water flow and pressure monitoring devices and fully completed the forelines for all the pumps. Just two weeks ago we evacuated the chambers for the first time. Within hours the chambers reached a pressure of 5×10^{-6} torr (and still falling) and maintained this pressure as the source chamber was rotated. With this test, the main and source chambers are fully complete; we are now concentrating on the completion of the detector chamber as well as small internal parts which go in the source and main chambers for forming the supersonic expansion and for analyzing the velocity distribution of the molecular beam.

The progress on the internal parts which go in the source and main chambers and on the detector region is as follows. The design for supersonic molecular beam nozzle assembly is complete and submitted to Central Shop. The design for the velocity analyzer of the molecular beam is also complete and submitted. We are presently acquiring and constructing the associated motor driver and time synchronization electronics which go with this velocity analyzer -- these will be complete by the end of the summer as will the simple assembly to inlet the molecular sample to be studied into the supersonic nozzle. The only big job remaining on the apparatus is the detector.

The detector involves differential pumping regions, a liquid nitrogen cooled insert with ionizer, a quadrupole, and a Daly detector. This modified design of the molecular beam apparatus involved the construction of very difficult differential regions which reduce the background in the detector substantially at 10° from the molecular beam. The design was submitted to Central Shop in the fall; since then these chambers have been completely roughed machined, stress relieved, measured in our laboratory for final machining of precision apertures, and then final machined. The only other tricky part of the apparatus is the ionizer insert. These designs are presently submitted to Central Shop and work will begin on them soon. Over the summer we will finish the easy design of the quadrupole region and Daly detector.

We project that the detector will be completed in Central Shop by the end of 1988 as we work concurrently on the last few parts of the design as outlined and on integrating the detector with the already finished portion of the machine. This involves an intermediate alignment of the centerline of the ionization insert at liquid nitrogen temperature before the final machining of that insert, assembly of the ionizer and Daly detector, as well as construction of detector pumping forelines and interlocks (as we have just completed for the source and main chamber) and support stand and rails to mount the detector so it may be baked out when isolated from the main chamber. We will work vigorously to try to complete the construction and assembly of the detector by December, after which we can finally incorporate the required data acquisition electronics and begin pursuing the first crucial experiments outlined in the proposal. The amount of construction and assembly work we will endeavor to complete between now and December corresponds to about 1/3 of the apparatus, the other 2/3 being complete and tested in our laboratory; so far the progress corresponds to that outlined as a goal in our initial proposal.

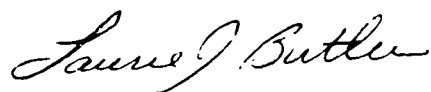


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The capabilities of the molecular beam time-of-flight techniques are unsurpassed for studying systems having many energetically allowed decomposition channels. While this construction effort involves a tremendous investment of time and money, the results obtainable with the apparatus are extremely important and can be obtained in no other way for these key systems. Thank you for your continuing support and please let me know if the form of this annual letter report is what you require, as this is the first I have written to you.

Yours sincerely,

A handwritten signature in cursive script, reading "Laurie J. Butler". The signature is fluid and elegant, with a prominent initial "L".

Laurie Butler

Enclosure

New Detector and Nozzle Drawings for Rotating Source Machine
Butler Group
University of Chicago, James Franck Institute

Detector/Differential	Differential Regions and Ionization	RSM-D-1-1E
"	Tee Outside Views	
"	Main Chamber Coupling Flange	RSM-D-1-2D
"	Differential Turbo Pump Ports	RSM-D-1-3B
"	Base Plate for Regions I & II	RSM-D-1-4C
"	Region II Wall A	RSM-D-1-5B
"	Region II Wall B	RSM-D-1-6A
"	Region II Wall C	RSM-D-1-7A
"	Coverplate - Region II	RSM-D-1-8B
"	Region I Wall A	RSM-D-1-9C
"	Region I Front Coverplate	RSM-D-1-10A
"	Region I Angled Final Coverplate	RSM-D-1-11A
"	Region I Wall B	RSM-D-1-12C
"	Crosssections of Differential Regions and Tee for Ionization Region	RSM-D-1-13D
"	Gate Valve	RSM-D-1-14A
"	Gate Valve Rails	RSM-D-1-15A
"	Final Machining Detail Regions I & II	RSM-D-1-16D
"	First Differential Wall Aperture	RSM-D-1-17A
"	Second Differential Wall Aperture	RSM-D-1-18A
Detector/Ionization		
Region	Tee for Ionization Region	RSM-D-2-1D
"	Insert	RSM-D-2-2D
"	LN ₂ Feed Tube	RSM-D-2-3C
"	Brinks Ionizer Retaining Plate	RSM-D-2-4A
"	Brinks Ionizer Filament Lead Holder	RSM-D-2-5A
"	Brinks Ionizer Repeller & Filament Lead	RSM-D-2-6A
"	Brinks Ionizer Ion Lenses	RSM-D-2-7A

"	Brinks Ionizer Holder Plate	RSM-D-2-8B
"	Brinks Ionizer Grid Spotwelding Template	RSM-D-2-9B
"	Brinks Ionizer Assembly and Mounting Option I	RSM-D-2-10B
"	Brinks Ionizer Grid Holder	RSM-D-2-11A
"	Vacuum Braze Detail for LN ₂ Feed Tube	RSM-D-2-12B
"	Vacuum Braze Detail for Ionizer Holder Plate	RSM-D-2-13B
"	Vacuum Braze Detail for Insert-2 Options	RSM-D-2-14C
Source/Nozzle Assembly		
"	Differential Region Skimmer for 3mm ² Collision Region	RSM-S-1-1A
"	Source Region Skimmer	RSM-S-1-2A
"	Self Aligning Nozzle Support Cage	RSM-S-1-3B
"	Sliding Key Between Nozzle and Support Cage	RSM-S-1-4A
"	Nozzle Tube with .005" hole	RSM-S-1-5B
"	Nozzle Clamp	RSM-S-1-6A
Time-of-Flight Wheel		
Modifications	T.O.F. Flange to Cylinder Mating Plate	RSM-T-1-1B
"	T.O.F. Height Adjust Mating Plate	RSM-T-1-2B
"	T.O.F. Motor Cooling Block	RSM-T-1-3B
"	T.O.F. Keyed Flange	RSM-T-1-4B
"	T.O.F. Motor Mount	RSM-T-1-5C